

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

0365-0510P

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

09/889496

INTERNATIONAL APPLICATION NO. / JCTES

PCT/FI00/00039

January 18, 2000

PRIORITY DATE CLAIMED

January 18, 1999

TITLE OF INVENTION

PAINT COMPOSITIONS

APPLICANT(S) FOR DO/EO/US

SALMI, Maarit; KOSKIMIES, Salme; AIROLA, Karri; JANSSEN, Robert

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau. WO 00/44836
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a. ☒ is transmitted herewith.
  - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4)
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 20. below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98./International Search Report (PCT/ISA/210) with PTO-1449 and 10 cited documents
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821-1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:
  - 1.) PCT Request (PCT/RO/101)
  - 2.) One (1) Sheet Formal Drawings
  - 3.) International Preliminary Examination Report (PCT/IPEA/409)

U.S. APPLICATION NO. of known, reg. 37 CFR 1.51 <b>09/889496</b>		INTERNATIONAL APPLICATION NO. PCT/FI00/00039		ATTORNEY'S DOCKET NUMBER 0365-0510P	
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<p>21. <input checked="" type="checkbox"/> The following fees are submitted:</p> <p><b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</b>          Neither international preliminary examination fee (37 CFR 1.482)          nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO          and International Search Report not prepared by the EPO or JPO. .... <b>\$1,000.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) not paid to          USPTO but International Search Report prepared by the EPO or JPO. .... <b>\$860.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO          but international search fee (37 CFR 1.445(a)(2)) paid to USPTO. .... <b>\$710.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) paid to USPTO          but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$690.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) paid to USPTO          and all claims satisfied provisions of PCT Article 33(1)-(4) ..... <b>\$100.00</b></p> <p><b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b></p> <p>Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30          months from the earliest claimed priority date (37 CFR 1.492(e)).</p> <table border="1" style="width:100%; border-collapse: collapse;"> <tr> <th style="width:20%;">CLAIMS</th> <th style="width:20%;">NUMBER FILED</th> <th style="width:20%;">NUMBER EXTRA</th> <th style="width:20%;">RATE</th> <th style="width:20%;"></th> </tr> <tr> <td>Total Claims</td> <td>19 - 20 =</td> <td>0</td> <td>X \$18.00</td> <td>\$ 0</td> </tr> <tr> <td>Independent Claims</td> <td>3 - 3 =</td> <td>0</td> <td>X \$80.00</td> <td>\$ 0</td> </tr> <tr> <td colspan="4">MULTIPLE DEPENDENT CLAIM(S) (if applicable) YES + \$270.00</td> <td>\$ 270.00</td> </tr> <tr> <td colspan="4"><b>TOTAL OF ABOVE CALCULATIONS =</b></td> <td><b>\$ 1400.00</b></td> </tr> </table> <p><input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are          reduced by 1/2.</p> <p style="text-align: right;"><b>SUBTOTAL = \$ 1400.00</b></p> <p>Processing fee of <b>\$130.00</b> for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30          months from the earliest claimed priority date (37 CFR 1.492(f)).</p> <p style="text-align: right;"><b>TOTAL NATIONAL FEE = \$ 1400.00</b></p> <p>Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be          accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). <b>\$40.00</b> per property +</p> <p style="text-align: right;"><b>TOTAL FEES ENCLOSED = \$ 1400.00</b></p> <table border="1" style="width:100%; border-collapse: collapse;"> <tr> <td style="width:60%;"></td> <td style="width:20%; text-align: center;">Amount to be refunded</td> <td style="width:20%; text-align: center;">\$</td> </tr> <tr> <td></td> <td style="text-align: center;">charged</td> <td style="text-align: center;">\$</td> </tr> </table>	CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		Total Claims	19 - 20 =	0	X \$18.00	\$ 0	Independent Claims	3 - 3 =	0	X \$80.00	\$ 0	MULTIPLE DEPENDENT CLAIM(S) (if applicable) YES + \$270.00				\$ 270.00	<b>TOTAL OF ABOVE CALCULATIONS =</b>				<b>\$ 1400.00</b>		Amount to be refunded	\$		charged	\$	<p><b>CALCULATIONS PTO USE ONLY</b></p>
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a. ☒ A check in the amount of \$ **1400.00** to cover the above fees is enclosed.

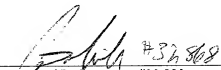
b. ☐ Please charge my Deposit Account. No. \_\_\_\_\_ in the amount of \$ \_\_\_\_\_ to cover the above fees.  
 A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any  
 overpayment to Deposit Account No. 02-2448.

**NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR  
 1.137(a) or (b)) must be filed and granted to restore the application to pending status.**

Send all correspondence to:  
**Birch, Stewart, Kolasch & Birch, LLP or Customer No. 2292**  
**P.O. Box 747**  
**Falls Church, VA 22040-0747**  
**(703)205-8000**

**Date: July 18, 2001**

By  #32,330  
 Leonard R. Svensson, #30,330

09/889496

JCIS Rec'd PCT/PTO 1 8 JUL 2001

PATENT  
0365-0510P

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicant: SALMI, Maarit et al  
Int'l. Appl. No.: PCT/FI00/00039  
Appl. No.: NEW Group:  
Filed: July 18, 2001 Examiner:  
For: PAINT COMPOSITIONS

PRELIMINARY AMENDMENT

**BOX PATENT APPLICATION**

Assistant Commissioner for Patents  
Washington, DC 20231

July 18, 2001

Sir:

The following Preliminary Amendments and Remarks are respectfully submitted in connection with the above-identified application.

AMENDMENTS

IN THE SPECIFICATION:

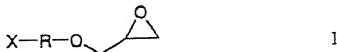
Please amend the specification as follows:

Before line 1, insert --This application is the national phase under 35 U.S.C. § 371 of PCT International Application No. PCT/FI00/00039 which has an International filing date of January 18, 2000, which designated the United States of America and was published in English.

**IN THE CLAIMS:**

Please amend the claims as follows:

6. (Amended) The paint composition according to claim 1, characterized in that it contains as a coalescent agent a glycidyl ether according to formula I



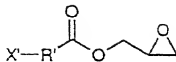
wherein R is a linear or branched, saturated or unsaturated

C<sub>3</sub>-C<sub>20</sub> hydrocarbon, optionally containing one or several hydroxyl groups, and

X represents hydrogen or a hydroxyl group.

7. (Amended) The paint composition according to claim 1, characterized in that the hydrocarbon residue of the glycidyl ether is derived from 1-butanol, 2-butanol, isobutanol, 1-pentanol, isopentanol, 1-hexanol, 2-ethylhexanol, 1-heptanol, 1-octanol, 2-ethyl-1,3-hexanediol, neopentyl glycol, 2-butyl-2-ethyl-1,3-propanediol, trimethylol ethane, trimethylol propane, 1,4-butanediol, neodecane alcohol, 1-6-hexanediol, 1,10-decanediol or 2-ethyl-2-hexen-1-ol.

8. (Amended) The paint composition according to claim 1, characterized in that it contains as a film forming agent a glycidyl ester according to formula II



II

wherein R' is a linear or branched, saturated or unsaturated C<sub>2</sub>-C<sub>20</sub> hydrocarbon, optionally containing one or several hydroxyl groups, and

X represents a methyl group, methylene hydroxy group or a carboxyl or lower carboxylate group.

9. (Amended) The paint composition according to claim 1, characterized in that the carboxylic acid residue of the glycidyl ester is derived from butanoic acid, isobutanoic acid, pentanoic acid, isopentanoic acid, 1-hexanoic acid, 2-ethylhexanoic acid, heptanoic acid, octanoic acid, neodecanoic acid, 2-hydroxyisobutanoic acid, 2,2-dimethyl-3-hydroxypropanoic acid, 2-ethyl-2-hexenoic acid, oleic acid, linolic acid, adipic acid, fumaric acid, maleic acid, succinic acid, glutaric acid or an anhydride or another derivative thereof.

10. (Amended) The paint composition according to claim 1, characterized in that the coalescent agent is 2-ethylhexylglycidyl ether, octyl glycidyl ether, mono- or diether of neopentylglycol or triglycidyl ether of trimetanolpropane, or 2-ethylhexyl glycidyl ester, octyl glycidyl ester or isopentyl glycidyl ester or methyl glycidyl ester of glutaric acid.

11. (Amended) The paint composition according to claim 1, characterized in that its pH is below about 8.5, preferably below 8.0.

12. (Amended) The paint composition according to claim 1, characterized in that it contains in addition to the glycidyl ether and/or ester at least one other coalescent agent, the proportion of the glycidyl ether and/or ester of the coalescent agents of the composition amounting to at least 20 wt.%, preferably at least 50 wt.%.

REMARKS

The specification has been amended to provide a cross-reference to the previously filed International Application.

The claims have been amended to delete the improper multiple dependencies and to place the application into better form prior to examination.

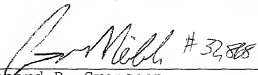
Entry of the above amendments is earnestly solicited. An early and favorable first action on the merits is earnestly solicited.

Attached hereto is a marked-up version of the changes made to the application by this Amendment.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17; particularly, extension of time fees.

Respectfully submitted,

BIRCH, STEWART, KOLASCH & BIRCH, LLP

By  #3288  
\_\_\_\_\_  
for Leonard R. Svensson  
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Attachments

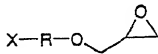
(Rev. 03/27/01)

VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS:

The claims have been amended as follows:

6. (Amended) The paint composition according to [any of claims 1 to 5] claim 1, characterized in that it contains as a coalescent agent a glycidyl ether according to formula I



I

wherein R is a linear or branched, saturated or unsaturated C<sub>3</sub>-C<sub>20</sub> hydrocarbon, optionally containing one or several hydroxyl groups, and X represents hydrogen or a hydroxyl group.

7. (Amended) The paint composition according to [any of the preceding claims] claim 1, characterized in that the hydrocarbon residue of the glycidyl ether is derived from 1-butanol, 2-butanol, isobutanol, 1-pentanol, isopentanol, 1-hexanol, 2-ethylhexanol, 1-heptanol, 1-octanol, 2-ethyl-1,3-hexanediol, neopentyl glycol, 2-butyl-2-ethyl-1,3-propanediol, trimethylol ethane, trimethylol propane, 1,4-butanediol, neodecane alcohol, 1-6-hexanediol, 1,10-decanediol or 2-ethyl-2-hexen-1-ol.



8. (Amended) The paint composition according to [any of claims 1 to 4] claim 1, characterized in that it contains as a film forming agent a glycidyl ester according to formula II



wherein R' is a linear or branched, saturated or unsaturated C<sub>2</sub>-C<sub>20</sub> hydrocarbon, optionally containing one or several hydroxyl groups, and  
X represents a methyl group, methylene hydroxy group or a carboxyl or lower carboxylate group.

9. (Amended) The paint composition according to [any of claims 1 to 5 or 8] claim 1, characterized in that the carboxylic acid residue of the glycidyl ester is derived from butanoic acid, isobutanoic acid, pentanoic acid, isopentanoic acid, 1-hexanoic acid, 2-ethylhexanoic acid, heptanoic acid, octanoic acid, neodecanoic acid, 2-hydroxy-isobutanoic acid, 2,2-dimethyl-3-hydroxypropanoic acid, 2-ethyl-2-hexenoic acid, oleic acid, linolic acid, adipic acid, fumaric acid, maleic acid, succinic acid, glutaric acid or an anhydride or another derivative thereof.

10. (Amended) The paint composition according to [any of the preceding claims] claim 1, characterized in that the coalescent agent is 2-ethylhexylglycidyl ether, octyl glycidyl ether, mono-

or diether of neopentylglycol or triglycidyl ether of trimetanolpropane, or 2-ethylhexyl glycidyl ester, octyl glycidyl ester or isopentyl glycidyl ester or methyl glycidyl ester of glutaric acid.

11. (Amended) The paint composition according to claim 1, characterized in that its pH is below about 8.5, preferably below 8.0.

12. (Amended) The paint composition according to [any of the preceding claims] claim 1, characterized in that it contains in addition to the glycidyl ether and/or ester at least one other coalescent agent, the proportion of the glycidyl ether and/or ester of the coalescent agents of the composition amounting to at least 20 wt.%, preferably at least 50 wt.%.

**Paint compositions**

The present invention concerns a paint composition according to the preamble of claim 1.

5 Generally, a composition of this kind contains a binder, a coalescent agent and admixtures and auxiliary agents known *per se*.

The invention further relates to a method according to the preamble of claim 14 for accelerating the hardening of a paint composition and to the use according to claim 16.

10 Many emulsion polymers used in latex paints do not form a film at moderate or low temperatures. In order to enable film formation at these conditions, film forming agents (*coalescent agents*) are added to latex paints. In solvent-borne paints the solvents provide for a lowering of the film formation temperature. The film forming chemicals are often organic solvents which evaporate into the surrounding air when the paint dries and hardens, thus giving rise to a smell and pollution problem. The traditional film forming agents therefore also increase the amounts of the volatile components (VOC) and the total emissions of the paint.

20 It is an aim of the development of coalescent agents to introduce on the market substituting coalescent agents which do not belong to the group of volatile organic compounds (VOC components). The disadvantage of non-volatile or slowly evaporating components is, however, their plasticising property, which gives rise to slow development of paint film hardness and which may leave the film in a soft condition.

25 Various reactive coalescent agents can be used in paint mixtures which should be kept free from emissions detrimental to the environment. These compounds should react with and form a part of the paint film, whereby the emissions minimized. The aim of using reactive coalescent agents is both to improve the development of the hardness and to reduce the

30 film formation temperature. These compounds are exemplified by, e.g., different isocyanates and epoxides and siloxanes.

Due to their structure the capability of reactive compounds of reducing the film formation temperature is often very small in comparison to normal volatile or so called no-VOC

35 coalescent agents. Furthermore, isocyanates are sensitive to moisture and cannot be used in

aqueous dispersions. In order to reach the film formation temperatures required for paints, conventional coalescent agents have to be used in addition to the reactive compounds.

It is an aim of the present invention to eliminate the disadvantages of the prior art and to provide entirely novel paint compositions.

The use of glycidyl esters in solvent-borne paints is known *per se*. Thus, published international patent application WO 97/44335 discloses that glycidyl esters can be used as reactive diluents for the preparation of epoxy, urethane and polyester paints. There is no mention in the publication about other properties of the glycidyl esters, e.g. their capability of forming films.

The present invention is based on the finding that glycidyl derivatives containing a large-sized hydrocarbyl residue, in particular glycidyl ethers and glycidyl esters, have excellent film formation properties and good dispersability in water. According to the invention the coalescent agents of water-borne paint compositions therefore comprise an ether or ester formed by glycidol (2,3-epoxy-1-propanol) and an organic alcohol or carboxylic acid containing 3 to 20 carbon atoms. Surprisingly it has been found that it is possible partially or totally to replace present coalescent agents, in particular reactive coalescent agents, of paint compositions. They make it possible considerably to accelerate the hardening of the paint film.

Glycidyl esters, such as the glycidyl ester of 2-ethylhexanol, can be prepared at high yield and in great purity from the corresponding carboxylic acid by first forming the alkali metal salt of the carboxylic acid, by mixing said salt with a molar excess of epichlorohydrin and by maintaining the reaction mixture at a temperature of at least 60 °C under reflux conditions. The reaction can be continued at said temperature without the formation of significant amounts of by-products, until the yield of the glycidyl ester is at least 80 %.

More specifically, the paint composition according to the present invention is characterized by what is stated in the characterizing part of claim 1.

The method of accelerating the hardening of a paint composition according to the present invention is characterized by what is stated in the characterizing part of claim 14, and the use according to the invention by what is stated in the characterizing part of claim 16.

The invention provides considerable advantages. Thus, the film forming agents according to the invention provide good dispersion of paint compositions which appears to be caused by the weakly polar ether bond or the more polar ester bond. Further, because of the rather large hydrocarbon group of the ether or ester, the novel coalescent agents reduce the film forming temperature better than conventionally used siloxanes or epoxy compounds. Due to the low volatility of the compounds they can be used in so called no-VOC applications. The reactive glycidyl group contained in the compounds makes it further possible to have the coalescent agents react so that they form a part of the paint film, whereby the total emissions are cut and there is an advantageous development of the hardness.

With the compounds it becomes possible to accelerate the hardening of the film so as to attain at least 70 % of the final hardness already within a day. The present glycidyl ethers or esters can be used together with known film forming agents.

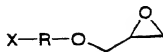
Next, the invention will be examined more closely with the aid of a detailed description and a number of working examples.

The attached drawing shows the development of König-pendulum hardness on glass sheets ( $t=23^{\circ}\text{C}$ ,  $rh=50\%$ ) of Primal E-2955+NPG diglycidyl ether at different temperatures and at a molar ratio between the epoxy and carboxylic acid groups of 0.5 to 1.0.

The present invention uses as a coalescent agent of paint compositions glycidyl ethers and/or esters which contain a linear or branched, saturated or unsaturated hydrocarbonyl residue having a hydrocarbonyl residue with 3 to 20 carbon atoms, usually 4 to 18 carbon atoms, preferably 4 to 12 carbon atoms, which is linked to the glycidyl group via an ether or ester bond. Short carbon chains give a more rapid development of the hardness and increase the final hardness of the film.

Thus, in the ethers used in the invention, at least one alcohol residue is derived from glycidol (2,3-epoxy-1-propanol), which forms an ether group with a linear or branched monoalcohol, diol, triol, tetraol or pentol having 3 to 20 carbon atoms.

The structure of the preferred glycidyl ethers is depicted in formula I



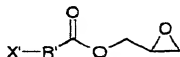
I

wherein R is a linear or branched, unsaturated or saturated C<sub>3</sub>-C<sub>20</sub> hydrocarbon, optionally containing one or several hydroxyl groups, and X represents hydrogen or a hydroxyl group.

The ether derivatives may contain 1, 2, 3, 4 or 5 glycidyl ether groups. Glycidyl ether groups may also be contained in glycidyl esters which are derived from hydroxyacids. Glycidyl ester compounds derived from diols and polyols may also contain ether groups derived from other alcohols (mixed ethers). In particular, the alcohols of these other alcohols are alkanols, such as lower alkanols (methanol, ethanol, propanol, butanol). The ether groups derived from other alcohols, should such compounds be present, form at the most 80 mole-%, preferably 1 to 50 mole-% of the ether groups of the ether-based coalescent agent.

The other main group of the present coalescent agents is formed by esters which have an alcohol residue derived from glycidol and a carboxylic acid residue derived from a linear or branched carboxylic acid comprising one or several protones (i.e. an acid which contains several carboxylic groups) and having 3 to 20, preferably 4 to 18, in particular 4 to 12 carbon atoms. The multibasic acids provide mono-, di-, tri- etc. esters. The esters can be semiesters or total esters.

The structure of preferred glycidylesters is depicted in formula II



II

wherein R' is a linear or branched, unsaturated or saturated C<sub>2</sub>-C<sub>20</sub>-hydrocarbon, which optionally contains one or several hydroxyl groups, and

X' represents a methyl group, a methylene hydroxy group or a carboxy or lower carboxylate group.

Examples of the lower carboxylate groups are, in particular, methoxylate (-COOMe) and ethoxylate (-COOEt).

The esters can also be prepared from hydroxy acids, as mentioned above, whereby they may contain ether groups. The glycidyl esters of multibasic acids may contain in addition to the ester group derived from glycidol ester groups derived also from other alcohols (mixed esters). As mentioned above in connection with the ethers, these other alcohols are in particular alkanols, such as lower alkanols, e.g. methanol, ethanol, propanols or butanols.

Examples of ethers derived from monoalcohols include 2-ethylhexyl glycidyl ether and octyl glycidyl ether. Ethers derived from dialcohols are represented by mono- and diethers of neopentylglycol and as examples of triethers the triglycidyl ether of trimetanolpropan can be mentioned.

As examples of glycidyl esters, 2-ethylhexyl glycidyl ester, octyl glycidyl ester and isopentyl glycidyl ester and as examples of mixed esters the methyl glycidyl ester of glutaric acid can be mentioned.

The glycidyl ethers can be prepared by the Williamson ether synthesis starting from epichlorohydrin and alcohols. The preparation of glycidyl ethers has been described in an article by Kida, T., Yokota, M., Masuyama, A. ja Okahara, M., in Synthesis 1993, 487-489. Since the reaction between epichlorohydrin and a hydroxy group releases hydrogen chloride, the etherification is preferably carried out in an alkaline medium which is capable of binding the hydrogen chloride. In the examples below a medium containing potassium hydroxide has been used, whereby the hydrogen chloride forms potassium chloride, which can be separated from the reaction system by filtration. As a solvent of the organic phase in the reaction system, an organic solvent, such as toluene, DMSO or an ether, such as THF, can be used.

The etherification is preferably carried out at normal air pressure and at a temperature in

the range of about 0 to 80 °C. Preferably the reaction is carried out at a temperature above room temperature (at above 25 °C) and the temperature is raised after the addition of the reactant in order to increase the reaction rate. The product is recovered from the organic phase by extraction and distillation at reduced pressure.

5

An excess of epichlorohydrin is used in relation to the number of hydroxyl groups. The molar ratio between the epichlorohydrin and the hydroxyl groups of the alcohol is 10:1 - 1:1, preferably 5:1 - 2:1.

10

As far as the invention is concerned, suitable alcohols for the preparation of reactive alkoxides are the below listed monoalcohols and diols and triols. Suitable alcohols are branched or linear monoalcohols, diols and triols comprising generally 3 to 20 (in particular 4 to 18, preferably 4 to 12) carbon atoms. Particularly suitable alcohols for use as starting materials of glycidyl ethers employed in paint compositions are, e.g., 1-butanol, 2-butanol, isobutanol, 1-pentanol, isopentanol, 1-hexanol, 2-ethylhexanol, 1-heptanol, 1-octanol, 2-ethyl-1,3-hexanediol, neopentylglycol, 2-butyl-2-ethyl-1,3-propanediol, trimethylolpropane and trimethylolpropane. Other suitable alcohols are 1,4-butanediol, neodecanalcohol, 1,6-hexanediol, 1,10-decanediol and 2-ethyl-2-hexen-1-ol.

20

The glycidyl esters according to the invention can be prepared from epichlorohydrin and acids and derivatives thereof e.g. by transesterification by reacting an alkali metal salt, e.g. a sodium salt, of a carboxylic acid with epichlorohydrin. For the dicarboxylic acid the starting materials can comprise, in addition to the acids, also the corresponding anhydride, from which a suitable alkali metal salt is formed. The preparation of glycidyl esters from carboxylic acids is disclosed in an article by Kester, E.B., Gaiser, C.J. and Lazar, M.E., in J. Org. Chem., 8 (1943) 550 and the preparation of alkyl glycidyl esters of anhydrides in an article by Zlatanos, S.N. and Sagredos, A.N., in JAOCS, 67 (1990) 661.

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The esters can also be prepared by direct esterification.

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The temperature of the actual esterification reaction is 30 to 120 °C, preferably about 50 to 100 °C. The reaction is carried out at normal air pressure. A catalyst can be used during esterification in an amount of 1 to 40 % of the amount of the esterification agent. Suitable esterification catalysts are p-toluenesulphonic acid, benzenesulphonic acid, sulphuric acid, tin and zinc salts or oxides and titanates. Suitable transesterification catalysts are alkali metal alkoxides, such as potassium or sodium alkoxides, sulphuric acid, hydrochloric acid

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and acid ion exchange resins.

According to a preferred embodiment the esterification is carried out by first forming an alkali metal salt from a linear or branched, unsaturated or saturated carboxylic acid having 3 to 20 carbon atoms. A reaction mixture is then formed from the alkali metal salt of the carboxylic acid and a molar excess of epichlorohydrin which is refluxed at a temperature of at least 60 °C. Since the reaction mixture contains several components, the reaction temperature can vary, depending on the conditions of the refluxing, even with 20 to 50 degrees. A typical temperature range during the reaction is about 65 to 100 °C. The reaction is continued after this for at least 1 hour at said temperature until the yield of the glycidyl ester is at least 80 %, typically at least 90 %. The formation of the by-products is less than 5 % of the end product, and the purity of the product is over 95 %. During the reaction the excess of epichlorohydrin amounts to at least 50 %, typically the molar ratio of epichlorohydrin to the number of carboxy groups of the reacting carboxylic acid is 2- to 5-fold. Surprisingly it has been found that the glycidyl ester of, e.g. 2-ethylhexanoic acid, can be produced at rather high yield, typically amounting to over 90 % and at high purity (purity in excess of 95 %).

Acids suitable for esterification comprise generally branched or linear mono- or dicarboxylic acids and hydroxy acids having 4 to 18 carbon atoms. Particularly suitable acids as starting compounds of glycidyl esters used in paint compositions are, e.g., butanoic acid, isobutanoic acid, pentanoic acid, isopentanoic acid, 1-hexanoic acid, 2-ethylhexanoic acid, heptanoic acid, octanoic acid, neodecanoic acid, 2-hydroxy-isobutanoic acid, 2,2-dimethyl-3-hydroxypropanoic acid, adipic acid, fumaric acid and maleic acid, succinic acid, glutaric acid and anhydrides and other derivatives thereof and 2-ethyl-2-hexenoic acid, oleic acid and linoleic acid.

The glycidyl ethers and glycidyl esters according to the invention can be formulated into water-borne dispersions, which contain a water-dispersible binder and admixtures and auxiliary agents known *per se*. The binding agents may comprise binders of latex paints, such as poly(vinyl acetate) and polyacrylates. Generally the polyacrylates comprise a synthetic latex, which is an ethylenically unsaturated polymer or copolymer, such as acrylic acid, itaconic acid or maleic acid. The poly(vinyl acetate) may include comonomers having carboxylic groups. Other binders which can be dispersed into water include various water-soluble polymers (e.g. PVA), starch, CMC, hydroxyethyl cellulose and poly(vinyl alcohol).

In addition to the above mentioned binding agents various water-dispersable alkyds may be used. These generally comprise a condensate product formed from a bivalent polyol and fatty acid or a natural oil. To facilitate dispersion or even dissolution in water they typically contain carboxylic acid groups.

It is particularly preferred to use the glycidyl ethers and/or esters according to the invention in paint compositions which contains a binder which forms a hard film when the paint has dried. Suitable binders are, e.g. acrylate latexes, wherein styrene is polymerised with one or several acrylate or methacrylate monomers. Latexes of this kind are represented by styrene-acrylate-latexes having carboxy functions, such as styrene-methylmethacrylate or styrene-butylacrylate latexes. Example 11 below discloses the use of the coalescent agents according to the invention with styrene-ethylacrylate and styrene-2-ethylhexylacrylate-methylmethacrylate. Other suitable latexes which may be mentioned include styrene-2-ethylhexyl acrylate-vinylacetate and styrene-2-ethylhexylacrylate-methylmethacrylate-n-butylacrylate.

The amount of the binder is generally 20 to 80 % of the dispersion. In mat ceiling and wall paints, as well as in water-borne anticorrosion paints, even smaller amounts can be used. Typically, in commercial latexes, the amount of the actual polymer is about 30 to 60 %, in particular about 50 %. Since paints may contain as little as only 10 % latex, the amount of polymeric binder in mat paints is only about 5 %. Thus, the paint compositions according to the present invention contain binder in an amount of 3 to 80 %, in particular 4 to 60 %.

The admixtures and auxiliary agents of the dispersions are exemplified by surfactants, dispersion agents, thickening agents, conservation agents/fungicides, antifoaming agents and fillers and pigments, such as calcium carbonate and titanium dioxide, and pH regulating agents, such as aqueous ammonia.

The amount of reactive coalescent agents according to the present invention in a dispersion is about 0.01 - 20 wt-%, preferably 0.1 - 15 wt-%. The coalescent agent may be constituted solely by the glycidyl ether, glycidyl ester or a mixture thereof or a mixture of ethers and/or esters and conventional coalescent agents. Generally the portion of glycidyl ethers or esters is at least 20 wt-%, typically over 50 % of the total amount of coalescent agents. Preferably the present glycidyl derivatives are used together with coalescent agents or mixtures thereof having a boiling point higher than 250 °C at normal air pressure. Coalescent agents of this kind are, e.g. phenyl ethers of ethylene glycol, monoisobutyrate

of 2,2,4-trimethyl-1,3-pentanediol monoisobutyraceti (supplied under the trade name Texanol), n-butylether acetate of diethyleneglycol and the mono-n-butyl ether of dipropyleneglycol and the mono-n-butylether of tripropylene glycol (supplied under the trade names Dowanol DpnB and Dowanol TPnB, respectively).

The present glycidyl derivatives are capable as such or in combination with known film forming agents considerably to accelerate hardness development of paint compositions. The final hardness is reached already within 7 to 14 days, whereby at least 70 %, in particular at least 80 % and usually at least 90 % of the final film hardness is obtained within 24 hours when at least 20 %, preferably at least 50 %, of the film forming agent of the composition is formed by the glycidyl derivative.

As regards stability of the present reactive film forming agents and as far as the use of the paint compositions is concerned, the pH of the paint compositions is preferably neutral or acid, in particular neutral or slightly acidic. In particular the pH of the paint compositions is below about 8.5, preferably below 8.0.

The present film forming agents have been compared with commercial products. In paint compositions, the glycidyl compounds of Examples 4, 5 and 9 are capable of reducing the film forming temperature and in this respect they work better than the commercial compounds (Coatosil 1770) used for reference. Tables 1 and 2 will show that the boiling points of many of the prepared glycidyl compounds are in excess of 250 degrees and their volatilization rates are low, whereby their use is safer than of conventional reactive monomers, due to low volatility. The hardness of the paint films develops advantageously for every tested glycidyl compound. Examples 11 studies the development of the König Hardness of the Primal E- 2955+NPG diglycidyl ether on a glass substrate at different temperatures at a molar ratio of the epoxy groups to the carboxy groups of 0.5 to 1.0. At all temperatures, the hardness developed better than that of the 0 samples.

The following non-limiting examples illustrate the invention.

**Example 1***Preparation of the glycidyl ether of 2-ethylhexanol*

3 molar equivalents of epichlorohydrin are slowly added to a solution of 1 mol-eq. 2-ethylhexanol and 2 mol-eq of potassium hydroxide in DMSO. The mixture is heated during the addition of epichlorohydrin at about 35 °C and after the addition at 45 °C. The formed KCl salt is filtered and the organic phase is extracted with ether. After drying, the product is distilled at reduced pressure. The purity of the product was 97 % and the yield after distillation 50 % of the theoretical yield.

**Example 2***Preparation of the glycidyl ether of 1-octanol*

3 molar equivalents of epichlorohydrin are slowly added to a solution of 1 mol-eq. 1-octanol and 2 mol-eq. potassium hydroxide in DMSO. The mixture is heated during the addition of epichlorohydrin at about 35 °C and after the addition at 45 °C. The formed KCl salt is filtered and the organic phase is extracted with ether. After drying the product is filtered at reduced pressure. The purity of the product was 100 % and the yield after distillation was 32 % of the theoretical yield.

**Example 3***Preparation of the glycidyl ether of isobutanol*

3 molar equivalents of epichlorohydrin are slowly added to a solution of 1 mol-eq. isobutanol and 2 mol-eq. potassium hydroxide in DMSO. The mixture is heated during the addition of epichlorohydrin at about 35 °C and after the addition at 45 °C. The formed KCl salt is filtered and the organic phase is extracted with ether. After drying the product is filtered at reduced pressure. The purity of the product was 94 % and the yield after distillation was 17 % of the theoretical yield.

**Example 4***Preparation of the diglycidyl ether of neopentylglycol*

3 molar equivalents of epichlorohydrin are slowly added to a solution of 1 mol-eq. neopentyl alcohol and 2 mol-eq. potassium hydroxide in DMSO. The mixture is heated during the addition of epichlorohydrin at about 35 °C and after the addition at 45 °C. The

formed KCl salt is filtered and the organic phase is extracted with ether. After drying the product is filtered at reduced pressure. The purity of the product was 97 % and the yield after distillation was 50 % of the theoretical yield.

5 *Table 1. Properties of the glycidyl ethers prepared in Examples 1 to 4*

Name	Bp (°C)	volatilization rate (ButAc = 1)
Example 1. 2-ethylhexyl glycidylether	221	0.005110
Example 2. octyl glycidylether	255	-
Example 3. isobutyl glycidylether	168	-
Example 4. neopentylglycol diglycidylether	275	0.000387

### Example 5

#### 10 *Glycidylester of 2-ethylhexyl acid*

From 1 mol-eq. 2-ethylhexanoic acid and 2 mol-eq. NaOH in toluene the sodium salt of the acid was prepared. The water formed was removed with the aid of the toluene by azeotropic distillation. Then the reaction mixture is cooled to 50 °C and 3 mol-eq.

- 15 epichlorohydrin are slowly added. After the addition of the epichlorohydrin the mixture was refluxed for 1 – 8 h at 65 to 100 °C. The epichlorohydrin is removed by distillation and the product is distilled at reduced pressure. The yield of the product after the reaction was 94 % and the purity of the distilled product was 98 %.

### 20 **Example 6**

#### *Glycidylester of octanoic acid*

From 1 mol-eq. octanoic acid and 2 mol-eq. NaOH in toluene the sodium salt of the acid was prepared. The water formed was removed with the aid of the toluene by azeotropic

- 25 distillation. Then the reaction mixture is cooled to 50 °C and 3 mol-eq. epichlorohydrin are slowly added. After the addition of the epichlorohydrin the mixture was refluxed for 1 – 8 h at 65 to 100 °C. The epichlorohydrin is removed by distillation and the product is distilled at reduced pressure. The yield of the product after the reaction was 24 % and the purity of the distilled product was 90 %.

**Example 7***Glycidylester of isobutanoic acid*

From 1 mol-eq. isobutanoic acid and 2 mol-eq. NaOH in toluene the sodium salt of the acid was prepared. The water formed was removed with the aid of the toluene by azeotropic distillation. Then the reaction mixture is cooled to 50 °C and 3 mol-eq. epichlorohydrin are slowly added. After the addition of the epichlorohydrin the mixture was refluxed for 1 – 8 h at 65 to 100 °C. The epichlorohydrin is removed by distillation and the product is distilled at reduced pressure. The yield of the product after the reaction was 65 % and the purity of the distilled product was 100 %.

**Example 8***Glycidylester of 2,2-dimethyl-3-hydroxypropanoic acid*

From 1 mol-eq. 2,2-dimethyl-3-hydroxypropanoic acid and 2 mol-eq. NaOH in toluene the sodium salt of the acid was prepared. The water formed was removed with the aid of the toluene by azeotropic distillation. Then the reaction mixture is cooled to 50 °C and 3 mol-eq. epichlorohydrin are slowly added. After the addition of the epichlorohydrin the mixture was refluxed for 1 – 8 h at 65 to 100 °C. The epichlorohydrin is removed by distillation and the product is distilled at reduced pressure. The yield of the product after the reaction was 45 % and the purity of the distilled product was 87 %.

**Example 9***Methyl glycidylester of glutaric acid*

1 molar equivalent glutaric anhydride is added to a 1 N solution of NaOH in methanol/toluene. The mixture is cooled and the excess methanol is removed by distillation. The obtained sodium salt of the glutaric acid methyl ester is recrystallized from acetone and dried. 1 molar equivalent of the sodium salt of the glutaric acid and epichlorohydrin are mixed together and refluxed for 1 to 3 hours at 100 to 115 °C. The catalyst may comprise tert-ethylammonium iodide in an amount of 0.02 - 0.1 molar equivalents of the sodium salt. The product mixture is cooled, filtered and the excess epichlorohydrin is removed by distillation. The cooled mixture is extracted with ether and distilled in vacuum. The yield of the product is 72 % and the purity 89 %.

**Example 10***Glycidylester of isopentanoic acid*

From 1 mol-eq. isopentanoic acid and 2 mol-eq. NaOH in toluene the sodium salt of the acid was prepared. The water formed was removed with the aid of the toluene by azeotropic distillation. Then the reaction mixture is cooled to 50 °C and 3 mol-eq. epichlorohydrin are slowly added. After the addition of the epichlorohydrin the mixture was refluxed for 1 – 8 h at 65 to 100 °C. The epichlorohydrin is removed by distillation and the product is distilled at reduced pressure. The yield of the product after the reaction was 77 % and the purity of the distilled product was 96 %.

The boiling point of the isopentanoic acid glycidylester is 226 to 227 °C.

**Table 2.** Properties of the glycidylesters of Examples 5 to 9

Name	Bp (°C)	volatilization rate (ButAc = 1)
Example 5. 2-ethylhexyl glycidylester	242-246	0.001984
Example 6. Octyl glycidylester	190	-
Example 7. Isobutyl glycidylester	69-71 / 0.1 mmHg	-
Example 8. 2,2-dimethyl-3-hydroxypropanoic acid glycidylester	-	-
Example 9. Glutaric acid methylglycidylester	282-283	0.000067

**Example 11**

*Use of the prepared glycidyl compounds as film forming agents in water-based colour compositions.*

The applicability of some of the film forming agents prepared according to Examples 1 to 9 in colour compositions were tested by producing from two different commercial latex dispersion (Joncryl 95 and Primal E-2955) mixtures which contained glycidyl compounds admixed at different concentrations. A commercial film forming agent, viz. epoxysiloxane

(Coatosil 1770) was used for reference. The impact of the various agents on the film forming temperature will appear from the attached table.

Table 3. The influence of the glycidyl compounds prepared in Examples 4, 5 and 9 on the film forming temperature of Joncryl-95<sup>®</sup> and Primal E-2955<sup>®</sup> latexes compared with that of a commercial reactive film forming agent (Coatosil 1770) (Joncryl 95 MFFT = 16 °C, dry substance 30 %, Primal E-2995 MFFT = 56 °C\*, dry substance 37 %)

	Joncryl-95 0%	Joncryl-95 3%	Joncryl-95 6%	Primal- 2955 0%	Primal- 2955 3%	Primal- 2955 6%	Primal- 2955 10%
Example 4. Diglycidylether of NPG	16	14	0	40	36	24	15
Example 5. Glycidylester of 2- EHA	16	10	0	40	32	19	11
Example 9. Meth. glycidylester of GA	16	14	0	40	37	28	16
Ref. sample: Coatosil 1770	16	14	10	40	39	33	29

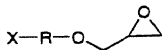
\* Value given by the manufacturer for the dry latex

The attached figure indicates how the König hardness of the Primal E-2955+NPG-dge sample has developed at different temperatures. As the figure shows, the hardness of a sample according to the invention is even up to three times as great as that of a 0 sample.



**Claims:**

1. Water-borne paint composition which comprises a binder, a coalescent agent and optionally admixtures and auxiliary agents known *per se*, characterized in that at least 20 % of the film forming agent is formed by a glycidyl ether or glycidyl ester or a mixture thereof, whereby the hydrocarbyl residue of the alcohol or carboxylic acid, respectively, of the ether or ester comprises a linear or branched, unsaturated or saturated hydrocarbon having 3 to 20 carbon atoms.
2. The paint composition according to claim 1, characterized in that it contains 0.01 to 20 wt.-% glycidyl ether and/or ester based on the dry matter content of the composition.
3. The paint composition according to claim 1 or claim 2, characterized in that it contains 3 to 80 wt.-%, in particular 4 to 60 wt.-% of a binder which can be dispersed in water.
4. The paint composition according to claim 3, characterized in that the binder is poly(vinyl acetate), polyacrylate, poly(vinyl alcohol), starch, carboxymethylcellulose, hydroxyethyl cellulose or alkyl.
5. The paint composition according to claim 4, characterized in that the binder forms a hard polymer film when the composition has dried.
6. The paint composition according to any of claims 1 to 5, characterized in that it contains as a coalescent agent a glycidyl ether according to formula I



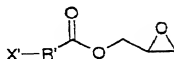
I

wherein R is a linear or branched, saturated or unsaturated C<sub>3</sub>-C<sub>20</sub> hydrocarbon, optionally containing one or several hydroxyl groups, and

X represents hydrogen or a hydroxyl group.

7. The paint composition according to any of the preceding claims, characterized in that the hydrocarbon residue of the glycidyl ether is derived from 1-butanol, 2-butanol, isobutanol, 1-pentanol, isopentanol, 1-hexanol, 2-ethylhexanol, 1-heptanol, 1-octanol, 2-ethyl-1,3-hexanediol, neopentyl glycol, 2-butyl-2-ethyl-1,3-propanediol, trimethylol ethane, trimethylol propane, 1,4-butanediol, neodecane alcohol, 1-6-hexanediol, 1,10-decanediol or 2-ethyl-2-hexen-1-ol.

8. The paint composition according to any of claims 1 to 4, characterized in that it contains as a film forming agent a glycidyl ester according to formula II



II

wherein R' is a linear or branched, saturated or unsaturated C<sub>2</sub>-C<sub>20</sub> hydrocarbon, optionally containing one or several hydroxyl groups, and

X represents a methyl group, methylene hydroxy group or a carboxyl or lower carboxylate group.

9. The paint composition according to any of claims 1 to 5 or 8, characterized in that the carboxylic acid residue of the glycidyl ester is derived from butanoic acid, isobutanoic acid, pentanoic acid, isopentanoic acid, 1-hexanoic acid, 2-ethylhexanoic acid, heptanoic acid, octanoic acid, neodecanoic acid, 2-hydroxy-isobutanoic acid, 2,2-dimethyl-3-hydroxypropanoic acid, 2-ethyl-2-hexenoic acid, oleic acid, linolic acid, adipic acid, fumaric acid, maleic acid, succinic acid, glutaric acid or an anhydride or another derivative thereof.

10. The paint composition according to any of the preceding claims, characterized in that the coalescent agent is 2-ethylhexylglycidyl ether, octyl glycidyl ether, mono- or diether of neopentylglycol or triglycidyl ether of trimetanolpropane, or 2-ethylhexyl

glycidyl ester, octyl glycidyl ester or isopentyl glycidyl ester or methyl glycidyl ester of glutaric acid.

11. The paint composition according to any of the preceding claims, characterized in that its pH is below about 8.5, preferably below 8.0.

12. The paint composition according to any of the preceding claims, characterized in that it contains in addition to the glycidyl ether and/or ester at least one other coalescent agent, the proportion of the glycidyl ether and/or ester of the coalescent agents of the composition amounting to at least 20 wt.-%, preferably at least 50 wt.-%.

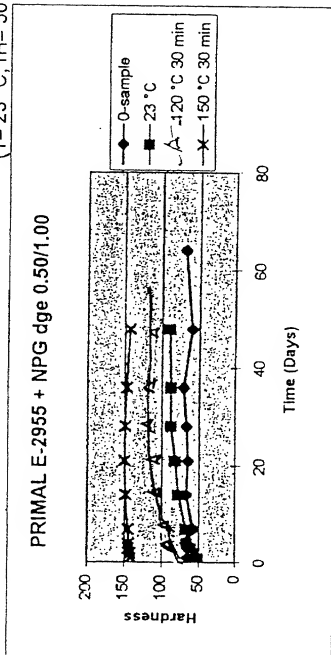
13. The paint composition according claim 12, characterized in that the other film forming agent is the phenyl ether of ethyleneglycol, 2,2,4-trimethyl-1,3-pentanediol monoisobutyrate and/or n-butyl ether acetate of diethyleneglycol.

14. Method of accelerating the hardening of an aqueous paint composition containing binder, coalescent agent and optionally admixtures and auxiliary agents known *per se*, characterized by incorporating into the composition a glycidyl ether or ester or a mixture thereof, wherein the hydrocarbyl residue of the alcohol or carboxylic acid, respectively, of the ether or ester comprises a linear or branched, unsaturated or saturated hydrocarbon having 3 to 20 carbon atoms, the proportion of the glycidyl ether and/or ester being at least 20 wt.-% of the film forming agents of the paint composition.

15. The method according to claim 14, characterized by incorporating at least 50 wt.-% of a glycidyl ether and/or ester, whereby the paint composition attains at least 70 % of its final hardness within a day.

16. Use of a glycidyl ether or ester as film forming agent in aqueous paint compositions, said ether or ester containing a linear or branched, saturated or unsaturated hydrocarbyl residue having 3 to 20 hydrocarbyl residue with 3 to 20 carbon atoms which is linked to the glycidyl group via an ether or ester bond.

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 $(T = 23\text{ }^{\circ}\text{C}, \text{rH} = 50\text{ } \%)$ .

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990089	Finland	January 18, 1999	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes <input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes <input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes <input type="checkbox"/> No
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Insert Citizenship

Insert Post Office Address

GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Maarit	Lahtinen	Maarit Lahtinen	July 16, 2001
Residence (City, State & Country)		CITIZENSHIP	
Jyväskylä, Finland <i>FIX</i>		Finnish	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			
Tikankolo 9 as 1, FIN-40520 Jyväskylä, Finland			
GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Kari	Rissanen	Kari Rissanen	July 16, 2001
Residence (City, State & Country)		CITIZENSHIP	
Hankasalmi, Finland <i>FIX</i>		Finnish	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			
Hankamäentie 13, FIN-41520 Hankasalmi, Finland			
GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Salme	Koskimies	Salme Koskimies	July 16, 2001
Residence (City, State & Country)		CITIZENSHIP	
Helsinki, Finland <i>FIX</i>		Finnish	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			
Hämeenapajantie 7, FIN-00850 Helsinki, Finland			
GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Karri	Airola	Karri Airola	July 16, 2001
Residence (City, State & Country)		CITIZENSHIP	
Porvoo, Finland <i>FIX</i>		Finnish	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			
Keltavuokontie 4 A 3, FIN-06400 Porvoo, Finland			
GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Robert	Jansson	Robert Jansson	July 16, 2001
Residence (City, State & Country)		CITIZENSHIP	
Helsinki, Finland <i>FIX</i>		Finnish	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			
Honkilahdentie 3, FIN-00940 Helsinki, Finland			